

AMENDMENTS TO THE CLAIMS

Please substitute the following claims 116-118, 120-128, 130, 139-145, 147, 148, 150-160, 162-166 and 184-195 as replacement claims for the previously-pending version of these claims. In this Supplemental Amendment C, claims 129, 131-138, 146 and 149 have been canceled, and claims 116, 123, 130, 141-144, 145, 147 and 148 have been amended.

1-115. (canceled).

116. (currently amended) A method for effecting chemical reactions of interest in ~~providing materials to a parallel processing microsystem for identifying and characterizing materials that enhance a chemical reaction,~~ the method comprising

simultaneously loading at least four catalyst materials into four or more microreactors such that the materials are individually resident in a reaction cavity of a separate microreactor, each of the at least four materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than about 3 ml for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity for supplying one or more reactants to the reaction cavity, and an outlet port in fluid communication with the reaction cavity for discharging a reactor effluent from the reaction cavity,

each of the four or more reactors being formed in a plurality of laminae, wherein the at least four catalyst materials are loaded into the four or more microreactors as a material-containing laminate comprising a substrate and the at least four catalyst materials at separate portions of the substrate,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

simultaneously unloading the catalyst-contacted materials from the microreactors in which they reside after the chemical reaction of interest by unloading the material-containing laminate.

117. **(original)** The method of claim 116 wherein at least ten materials are simultaneously loaded into ten or more microreactors.

118. **(original)** The method of claim 116 wherein at least one-hundred materials are simultaneously loaded into one-hundred or more microreactors.

119. **(canceled).**

120. **(previously presented)** The method of claim 116 wherein the plurality of laminae in which the four or more microreactors are formed include

the material-containing laminate as a material-containing first laminate comprising first and second surfaces in spaced, substantially parallel relationship to each other and the at least four materials,

a second laminate or composite substructure comprising a plurality of laminates, the second laminate or composite substructure comprising first and second surfaces in spaced, substantially parallel relationship with each other, and an array of four or more substantially coplanar wells arranged to correspond to the arrangement of the at least four materials of the first laminate, each of the wells having an interior surface defining an open-ended cavity at the first surface of the second laminate or composite substructure, the method further comprising

engaging the second surface of the material-containing first laminate and the first surface of the second laminate or composite substructure such that, taken together, the engaged laminae form an array of four or more microreactors defined by the interior surfaces of the wells and at least a portion of material-containing regions of the material-containing first laminate.

121. **(previously presented)** The method of claim 120 wherein the second surface of the material-containing first laminate and the first surface of second laminate are releasably engaged.

122. **(previously presented)** The method of claim 120 wherein the second surface of the material-containing first laminate and the first surface of second laminate are bonded.

123. **(currently amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

- simultaneously loading at least four candidate catalyst materials into four or more microreactors of a chemical processing microsystem such that the at least four materials are individually resident in separate microreactors, thereby forming four or more material-containing microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml, each of the four or more reactors being formed in a plurality of laminae, the at least four catalyst materials are loaded into the four or more microreactors as a material-containing laminate comprising a substrate and the at least four catalyst materials at separate portions of the substrate,
- simultaneously supplying one or more reactants to each of the four or more microreactors,
- simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,
- simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors,
- evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest, and
- simultaneously unloading the reactant-contacted materials from the microreactors in which they reside after the chemical reaction of interest by unloading the material-containing laminate.

124. **(original)** The method of claim 123 wherein the amount of each of the at least four materials loaded into the microreactors is not more than about 5 mg.

125. **(original)** The method of claim 123 wherein the amount of each of the at least four materials loaded into the microreactors is not more than about 1 mg.

126. **(original)** The method of claim 123 wherein at least ten materials are simultaneously loaded into ten or more microreactors.

127. **(original)** The method of claim 123 wherein at least one-hundred materials are simultaneously loaded into one-hundred or more microreactors.

128. **(original)** The method of claim 123 wherein the at least four materials are simultaneously loaded into the four or more microreactors as an array of candidate materials, the array comprising a substantially planar substrate and four or more materials at separate portions of the substrate.

129. **(canceled).**

130. **(currently amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

simultaneously loading at least four candidate catalyst materials into four or more microreactors formed in a plurality of laminae such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest, and

simultaneously unloading the reactant-contacted materials from the microreactors in which they reside after the chemical reaction of interest,

the at least four candidate catalyst materials being loaded into the four or more microreactors formed in the plurality of laminae, and the reactant-contacted materials being unloaded from the microreactors formed in the plurality of laminae, in each case without affecting the structural integrity of a fluid distribution system through which the one or more reactants are supplied to the microreactors or through which one or more reactor effluents are discharged from the microreactors.

131-138. (canceled)

139. (previously presented) A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a geometry and having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors,

selecting the geometry of the reaction cavity and controlling the reaction conditions in the four or more microreactors such that the reactant residence time, τ_{res} , is longer than the diffusion period, τ_{diff} , for the reaction cavity,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors, and

evaluating the at least candidate catalyst four materials for catalytic activity for the chemical reaction of interest.

140. **(previously presented)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least four candidate catalyst materials into four or more microreactors of a chemical processing microsystem such that the at least four materials are individually resident in separate microreactors, each of the at least four candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the four or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 10 μ l,

simultaneously supplying one or more reactants to each of the four or more microreactors,

simultaneously contacting each of the at least four candidate catalyst materials with the one or more reactants in the four or more microreactors,

simultaneously discharging a reactor effluent from each of the four or more material-containing microreactors into separate four or more microseparators of the chemical processing microsystem,

simultaneously separating one or more components of the reactor effluents in the four or more microseparators, and

evaluating the at least four candidate catalyst materials for catalytic activity for the chemical reaction of interest.

141. **(currently amended)** The method of claims 116, 123, 130, ~~131~~, 139 or 140 wherein the at least four materials are at least four different materials.

142. **(currently amended)** The method of claims 116, 123, 130, ~~131~~, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 100 μ l.

143. **(currently amended)** The method of claims 116, 123, 130, ~~131~~, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 10 μ l.

144. **(currently amended)** The method of claims 116, 123, 130, ~~131~~, 139 or 140 wherein the reaction cavity of each of the four or more material-containing microreactors has a volume of not more than about 1 μ l.

145. **(currently amended)** A method for identifying or optimizing catalysts for a chemical reaction of interest, the method comprising

loading at least twenty five candidate catalyst materials into twenty five or more microreactors using an automated material handling system such that the at least twenty five materials are individually resident in separate microreactors, each of the at least twenty five candidate catalyst materials comprising an inorganic material, a metal-ligand or a non-biological organic material, each of the twenty five or more microreactors comprising a surface defining a reaction cavity having a volume of not more than 3 ml,

simultaneously supplying one or more reactants to each of the twenty five or more microreactors,

simultaneously contacting each of the at least twenty five candidate catalyst materials with the one or more reactants in the twenty five or more microreactors under reaction conditions for the reaction of interest,

simultaneously discharging a reactor effluent from each of the twenty five or more material-containing microreactors, and

evaluating the at least twenty five candidate catalyst materials for catalytic activity for the chemical reaction of interest,

unloading the at least twenty five reactant-contacted materials from the microreactors in which they reside using the automated material-handling system, and

loading a second set of at least twenty five ~~four~~ materials into the twenty five ~~four~~ or more microreactors of the chemical processing microsystem using the automated material handling system such that the second set of at least twenty five ~~four~~ materials are individually resident in separate microreactors.

146. **(canceled).**

147. **(currently amended)** The method of claim ~~145~~ 146 wherein the at least twenty five candidate materials are loaded simultaneously into the twenty five or more microreactors, and the reactant-contacted candidate materials are unloaded simultaneously therefrom.

148. **(currently amended)** The method of claim ~~145~~ 146 wherein the at least twenty five materials are loaded sequentially into the twenty five or more microreactors, and the reactant-contacted candidate materials are unloaded sequentially therefrom.

149. **(canceled)**.

150. **(original)** The method of claim 145 wherein the one or more reactants are gaseous reactants.

151. **(previously presented)** The method of claim 145 wherein the at least twenty five materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be substantially the same in each of the twenty five or more microreactors.

152. **(previously presented)** The method of claim 145 wherein the at least twenty five candidate materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be substantially the same in at least a subset of the twenty five or more microreactors according to one or more control protocols selected from the group consisting of:

controlling the temperature to be not less than about 100 °C and to be substantially the same in at least four of the twenty-five or more microreactors,

controlling the pressure to range from about 1 atm to about 200 bar and to be substantially the same in at least four of the twenty five or more microreactors,

controlling the residence time to range from about 1 μ sec to about 1 hour and to be substantially the same in at least four of the twenty five or more microreactors, and

controlling the reactant flow rate to be substantially the same through at least four of the twenty five or more microreactors.

153. **(previously presented)** The method of claim 145 wherein the at least twenty five materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be varied between at least two of the twenty five or more microreactors.

154. **(previously presented)** The method of claim 145 wherein the at least twenty five materials are contacted with the one or more reactants under a set of reaction conditions, the method further comprising controlling the reaction conditions to be varied between at least two of the twenty five or more microreactors according to one or more control protocols selected from the group consisting of:

controlling the temperature to be varied between at least two of the twenty five or more microreactors,

controlling the pressure to be varied between at least two of the twenty five or more microreactors,

controlling the residence time to be varied between at least two of the twenty five or more microreactors, and

controlling the reactant flow rate to be varied through at least two of the twenty five or more microreactors.

155. **(original)** The method of claim 145 wherein the candidate materials are a film of material formed on a surface of the reaction cavity.

156. **(original)** The method of claim 145 wherein the microsystem comprises four-hundred or more microreactors.

157. **(original)** The method of claim 145 wherein the microsystem comprises one-thousand or more microreactors.

158. **(previously presented)** The method of claim 145 wherein different candidate materials are individually resident in the separate reaction cavities of at least 90% of the twenty five or more microreactors.

159. **(previously presented)** The method of claim 145 wherein at least twenty five materials are evaluated for catalytic activity according to one or more analytical protocols selected from the group consisting of

- determining catalytic activity by analytical measurement of the reactor effluent,
- determining catalytic activity by *in situ* analytical measurement,
- determining catalytic activity by serial analytical measurement,
- determining catalytic activity by parallel analytical measurement, and
- determining catalytic activity of a subset of the at least four materials by parallel analytical measurement.

160. **(previously presented)** The method of claim 145 wherein at least twenty five materials are evaluated for catalytic activity according to one or more analytical protocols selected from the group consisting of

- determining catalytic activity by parallel or serial gas chromatography of the reactor effluents,

- determining catalytic activity by separating one or more components of the reactor effluents and determining the presence, absence or amount of the separated one or more components,

- determining catalytic activity by adsorbing one or more components of at least four reactor effluents onto an adsorbent material, and determining the presence, absence or amount of adsorbed component,

- determining catalytic activity by adsorbing one or more components of at least four reactor effluents onto an adsorbent material, desorbing an adsorbed component, and determining the presence, absence or amount of desorbed component, and

- determining catalytic activity by determining the amount of a reaction product formed by the chemical reaction of interest.

161. (canceled).

162. (previously presented) A method for evaluating or optimizing process conditions for a chemical reaction of interest, the method comprising

- simultaneously supplying one or more reactants to each of four or more microreactors, each of the microreactors comprising a surface defining a reaction cavity having a volume of not more than about 10 μ l for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity, and an outlet port in fluid communication with the reaction cavity,
- controlling a first reaction condition to be substantially identical in each of the microreactors,
- controlling a second reaction condition to be varied between two or more of the microreactors,
- simultaneously discharging a reactor effluent from each of the four or more microreactors to four or more microseparators, each of the microseparators comprising a surface defining a separation cavity for separating at least one component of a reactor effluent, an inlet port in fluid communication with the outlet port of one of the microreactors for receiving the reactor effluent therefrom, and an outlet port in fluid communication with the separation cavity, and
- simultaneously discharging the separated effluent from each of the microseparators, and evaluating the effect of varying the second set of reaction conditions.

163. (original) The method of claim 162 wherein the four or more microreactors are formed in a plurality of laminae and the four or more microseparators are formed in a plurality of laminae.

164. (original) The method of claim 162 wherein the reaction cavity of each of the at least four candidate material-containing microreactors has a volume of not more than about 1 μ l.

165. (previously presented) The method of claim 162 wherein the first and second reaction conditions are independently selected from the group consisting of temperature, pressure, residence time and flow rate.

166. **(previously presented)** The method of claim 162 wherein the first and second reaction conditions are independently selected from the group consisting of temperature, pressure, and residence time.

167-183. **(canceled)**.

184. **(original)** A method for effecting a microscale chemical reaction, the method comprising

supplying one or more reactants for a chemical reaction of interest to a microreactor, the microreactor comprising a surface defining a reaction cavity having a volume of not more than about 10 μl for carrying out a chemical reaction, an inlet port in fluid communication with the reaction cavity for supplying one or more reactants thereto, and an outlet port in fluid communication with the reaction cavity for discharging one or more reaction products therefrom, and

converting the one or more reactants to one or more reaction products in the reaction cavity,

the reactants residing in the reaction cavity under process conditions effective for the chemical reaction of interest for a residence time, τ_{res} , that is longer than the diffusion period, τ_{diff} , for the reaction cavity under such process conditions.

185. **(original)** The method of claim 184 wherein the microreactor further comprises a catalyst within the reaction cavity for catalyzing the chemical reaction of interest.

186. **(previously presented)** The method of claim 123 wherein the one or more reactants are simultaneously supplied to each of the four or more microreactors through a fluid distribution system, the fluid distribution system providing fluid communication from one or more reactant sources to the reaction cavities of each of the four or more microreactors through a microfluidic fluid-supply manifold.

187. **(previously presented)** The method of claims 123 or 186 wherein the reactor effluent is simultaneously discharged from each of the four or more material-containing microreactors through an effluent distribution system providing fluid communication from the reaction cavity of each of the four or more microreactors to one or more effluent sinks through a microfluidic effluent-distribution manifold.

188. **(previously presented)** The method of claim 186 wherein the fluid distribution system is effective for supplying one or more gaseous reactants through the microfluidic fluid-supply manifold.

189. **(previously presented)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to be above 100 °C during the chemical reaction of interest.

190. **(previously presented)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to be above 200 °C during the chemical reaction of interest.

191. **(previously presented)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to range from about 100 °C to about 500 °C during the chemical reaction of interest.

192. **(previously presented)** The method of claim 123 further comprising controlling the temperature of the reaction cavities of each of the four or more microreactors to range from about 100 °C to about 800 °C during the chemical reaction of interest.

193. **(previously presented)** The method of claim 145 wherein the automated material handling system is adapted for simultaneously loading candidate catalyst materials into the microreactors.

194. **(previously presented)** The method of claim 145 wherein the automated material handling system is adapted for serially loading candidate catalyst materials into the microreactors.

195. **(previously presented)** The method of claim 145 wherein the automated material handling system is adapted for automatically or semiautomatically loading candidate catalyst materials into the microreactors.